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Continuous Working-Level Measurements Using Alpha or Beta Detectors

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CONTINUOUS WORKING-LEVEL MEASUREMENTS USING ALPHA OR BETA DETECTORS

by

Robert F. Drouillard¹ and Robert F. Holub²

ABSTRACT

The Bureau of Mines has investigated techniques of using gross alpha or beta detectors to continuously measure working levels. Both methods measure radioactive particulates collected on a filter paper using a constant airflow. Inherent-error studies indicate a value of about ± 3 percent for the gross alpha method and about ± 8 percent for the beta method in typical mine atmospheres. However, the beta method avoids problems associated with alpha detectors and is therefore more useful.

Applications of these continuous working-level detectors include work area monitoring of exposure levels in underground openings, such as mines and caves, and calibrating personal dosimeters exposed over extended time intervals.

INTRODUCTION

Exposure to airborne radionuclides is potentially hazardous to the respiratory tract and may result in lung cancer. The problem is particularly acute among underground miners in the uranium industry where daughter products of radon-222 are always present in the mine atmosphere. RaA and RaC' are of the most concern because of the tissue damage caused by absorption of the highly energetic alpha particles emitted during the decay of these polonium isotopes. There is no known method for accurately measuring this dose to the lung tissue; consequently, the miners' exposure to airborne radionuclides is measured instead. These exposure measurements are used for both recordkeeping and control of the radioactivity levels in the mine atmosphere.

Exposure levels in the domestic mining industry are based on the working level (WL), which is any combination of the short-lived radon daughter products in a liter of air that will result in the ultimate emission by them of

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1.3×10^5 mev of alpha energy.³ Evans (1)⁴ has reviewed the fundamental physical principles of the working level as well as other aspects of measuring filterable airborne radionuclides.

Current methods of measuring working-level exposures in underground mines are based on the collection of filterable radionuclides for a sample period, which is commonly 5 minutes. The frequency of these measurements at worksites in mines varies from mine to mine; some mines check worksites daily, while others may sample much less frequently.

For the past several years, the Bureau of Mines and the Mining Enforcement and Safety Administration have been studying improved methods of measuring working-level exposures. A large part of this effort has been on alpha dosimeters, with particular emphasis on the thermoluminescence dosimeter (TLD) to be used as a personal exposure monitor. During the course of testing the TLD devices in the laboratory and in mines, the need for a continuous working-level measuring technique to eliminate the need for frequent grab samples became evident. A review of the literature indicated two approaches that had been tried. Haider and Jacobi (3) reported on a continuous alpha-monitoring device that was used for 1 year in a fluorspar mine in eastern Bavaria, Germany. A diode-type alpha detector was used to measure alpha activity collected on a filter paper. In another study, Holmgren (4) used a commercial air particulate monitor and measured the beta activity of RaB + RaC to arrive at working-level exposure values.

The Bureau of Mines investigation covers the use of gross alpha and beta detectors as a means of continuous measurement of working levels with attention given to the inherent errors of these nonexact methods. Both methods of detection require continuous sampling of air at a known flow rate. The continuous air sampling of a given air concentration results in radioactive levels on the filter not found for the commonly used 5-minute sampling period. A comprehensive review of the growth and decay of radionuclides on a filter under continuous air sampling is provided by Evans (1).

DESCRIPTION OF THE DETECTORS

Beta Detector

Figure 1 shows a prototype unit using a 47-mm filter holder with a pancake-type Geiger-Mueller (GM) tube serving as a beta detector. The tube has a window with an areal density of about 9 mg/cm^2 to provide sufficient strength for operation under negative differential pressure conditions. The filter paper and the window of the GM tube are about 1.25 inches apart. The filter holder is a modified commercial unit which has been adapted to the aluminum housing. An air pump with controlled airflow is connected to the

³A value for the total alpha energy of 1.27800×10^5 mev is used in calculations for this report. "Alpha" and "beta" measurements denote particle measurements.

⁴Underlined numbers in parentheses refer to items in the list of references at the end of this report.

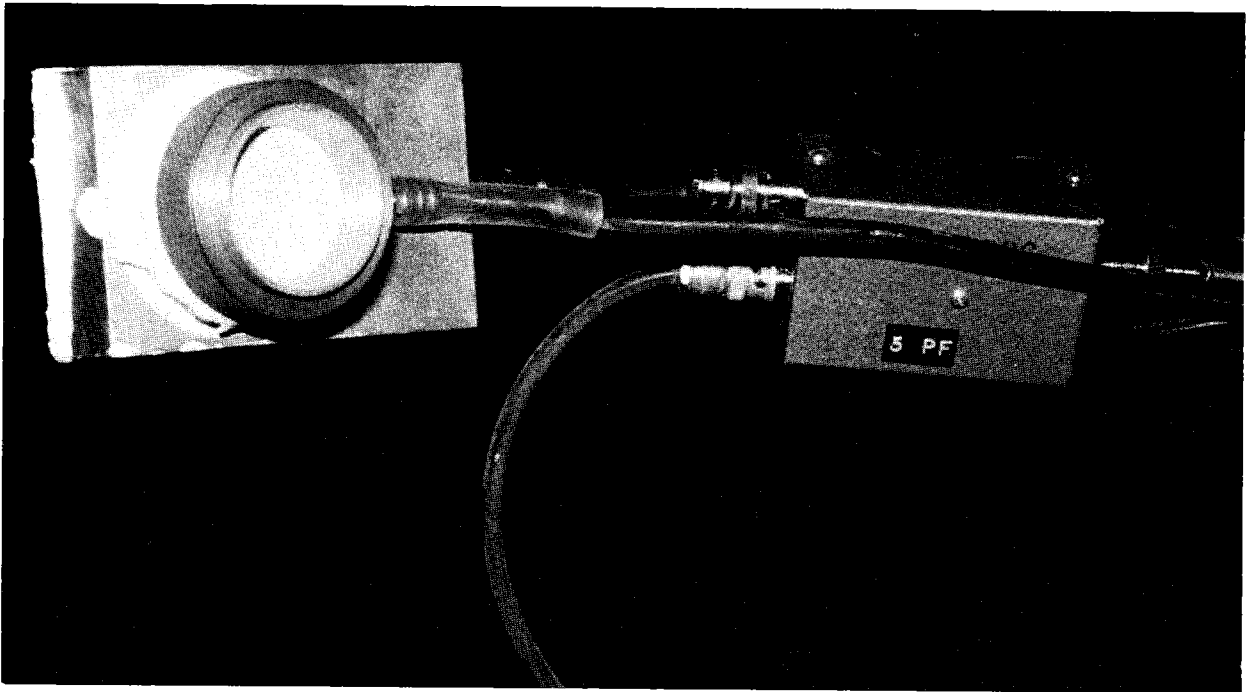


FIGURE 1. - Beta detector assembly.

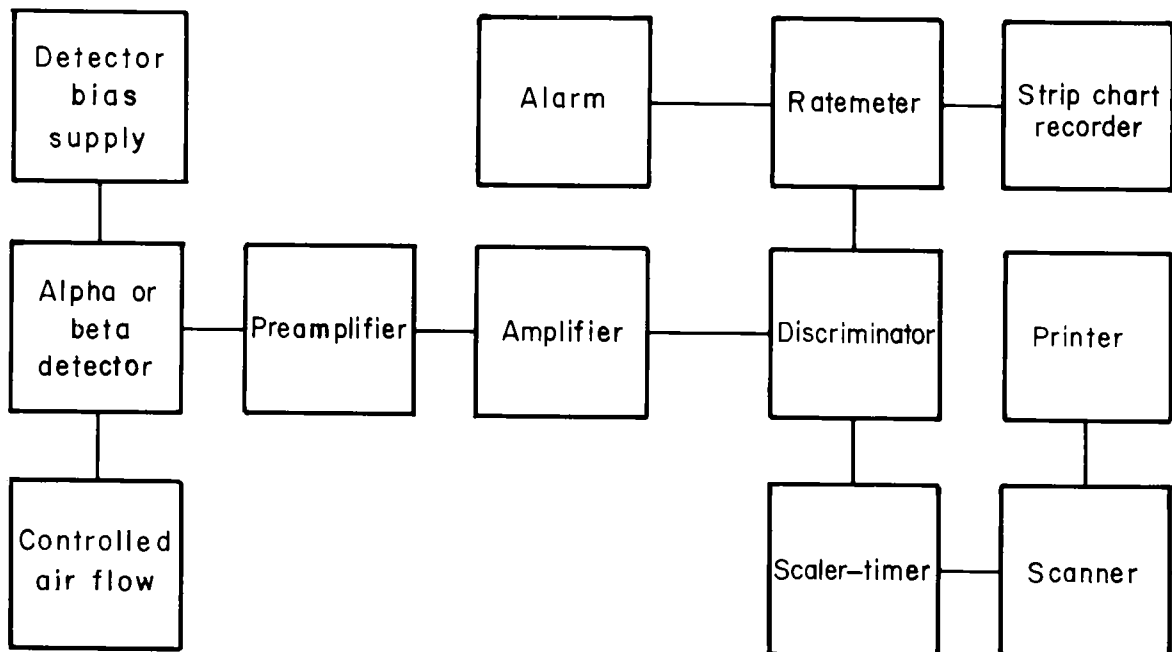


FIGURE 2. - Block diagram of typical counting system using either an alpha or a beta detector.

side of the filter holder. The airflow regulator is adjusted for a flow rate of 1.0 liter per minute, and regulation is provided over a differential pressure operating range of 0.5 to 40 inches water column.

The GM tube is operated at the manufacturer's specified voltage and may be coupled directly to portable scaler-timers, which are commercially available, or to a suitable preamplifier that can be used to drive remote counting equipment. Figure 2 is a block diagram of a typical counting system assembled from commercial units. The preamplifier and amplifier provide signal conditioning prior to a discriminator which provides logic pulses for the counting units. The scaler-timer, along with the scanner and printer, provides the digital data. Analog data output for display on a strip chart recorder is provided by a ratemeter. This system component also drives an alarm module. All of the aforementioned system components, except the preamplifiers and the data recorders, are standard nuclear instrument modules.

The beta detector also responds to gamma rays, and this background activity must be subtracted from the gross activity. No attempt has been made to provide background shielding, in the interest of holding the size and weight of the detector assembly to a minimum.

Alpha Detector

Figure 3 shows a prototype alpha detector-filter holder combination. The air to be sampled enters the housing through 18 holes, and the particulates are collected on the filter, which is located about 0.2 inch from the detector

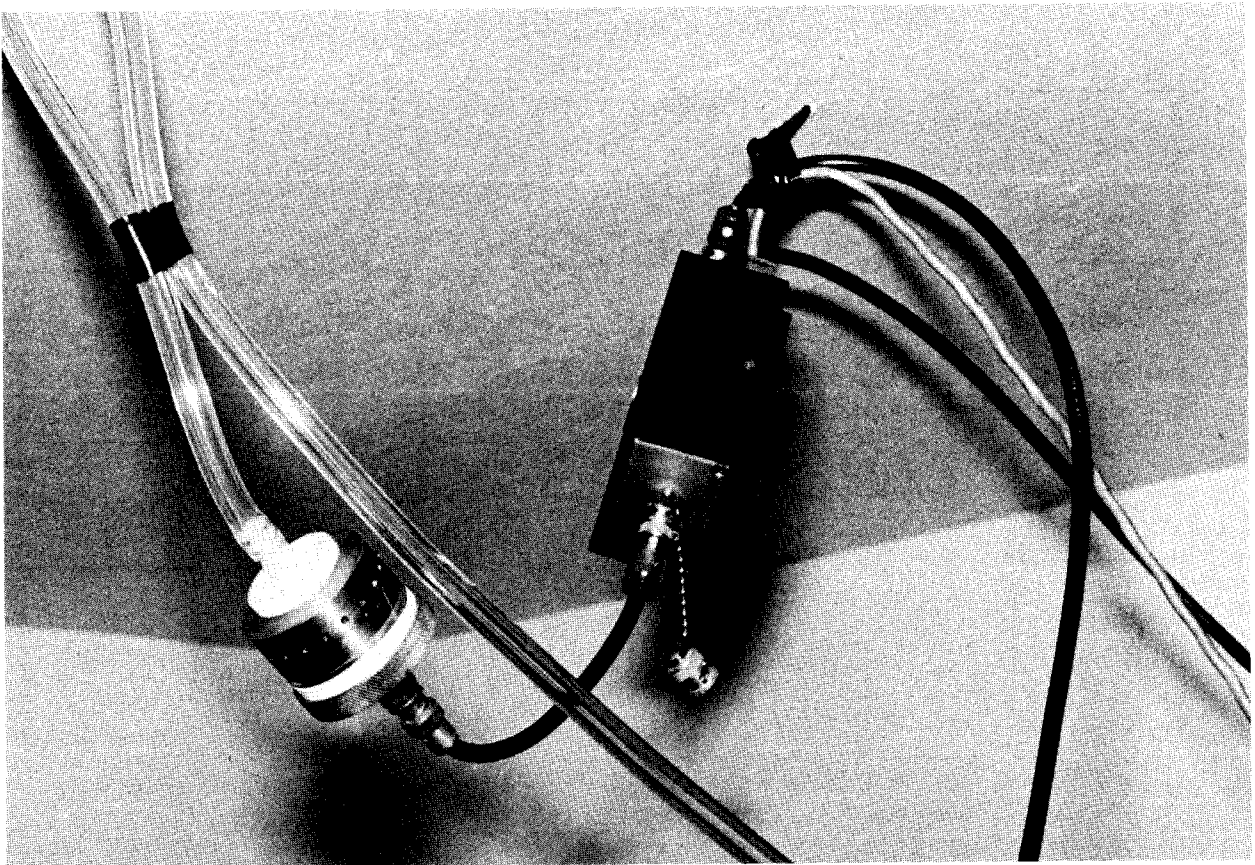


FIGURE 3. - Alpha detector assembly.

A surface-barrier-type diode detector, with an area of 300 mm², is used to measure the alpha particle activity on a 25-mm filter. An aluminized Mylar⁵ window separates the detector from the filter to provide some protection for the detector from moisture encountered in mine atmospheres.

The surface barrier detector is operated at the manufacturer's recommended bias voltage, and its output pulses are connected to a preamplifier for transmission by coaxial cable to the counting system. A typical counting system for this detector is the same as the one shown in figure 2 except for the preamplifier.

INHERENT ERROR ANALYSIS

The continuous beta and continuous gross alpha methods of measuring working levels are nonexact methods because they do not measure all of the components comprising a working level. This is also true of the Kusnetz method (2, 6), which is the industry standard, and of some of the "instant" working-level meters (8-9). It is essential, then, to evaluate inherent errors with respect to all possible mixtures of RaA, RaB, and RaC.

A triangular graph shown in figures 4 and 5 is a very useful way of displaying the distribution of mixtures of RaA, RaB, and RaC found in underground mine atmospheres and the inherent error for any nonexact method of measuring working levels. Any point within the triangle represents an airborne activity mixture of radon daughter products normalized to one working level. The lower left corner represents pure RaA with an activity of 956.3 picocuries, the upper right corner represents pure RaB with an activity of 193.9 picocuries, and the upper left corner represents pure RaC with an activity of 263.7 picocuries. Zero activity for RaA and RaB is at the upper left corner, while zero activity for RaC lies on the hypotenuse side of the triangle as shown in figure 4. For simplicity, the RaC axis is not shown in figure 5.

In all nonexact methods of measuring working levels, an effort is made to select a calibration that has the minimum inherent error over the widest range of daughter product mixtures commonly found in the environment being sampled. The graph in figure 5 shows the distribution of over 400 measurements of RaA, RaB, and RaC made in underground mines by a number of investigators. By overlaying this collection of data points with isoerror lines calculated for the conditions of measurement, the probable inherent error accuracy of various nonexact methods can be determined.

The distribution of radon daughter product mixtures measured in the Bureau of Mines radon test chamber lies within the central portion of most of the field data. Accordingly, this region of the distributions has been selected for the zero inherent error line, and a typical mixture found in the chamber is used to calculate isoerror lines for both the beta and the gross alpha methods of continuous working-level measurements.

⁵Reference to specific equipment does not imply endorsement by the Bureau of Mines.

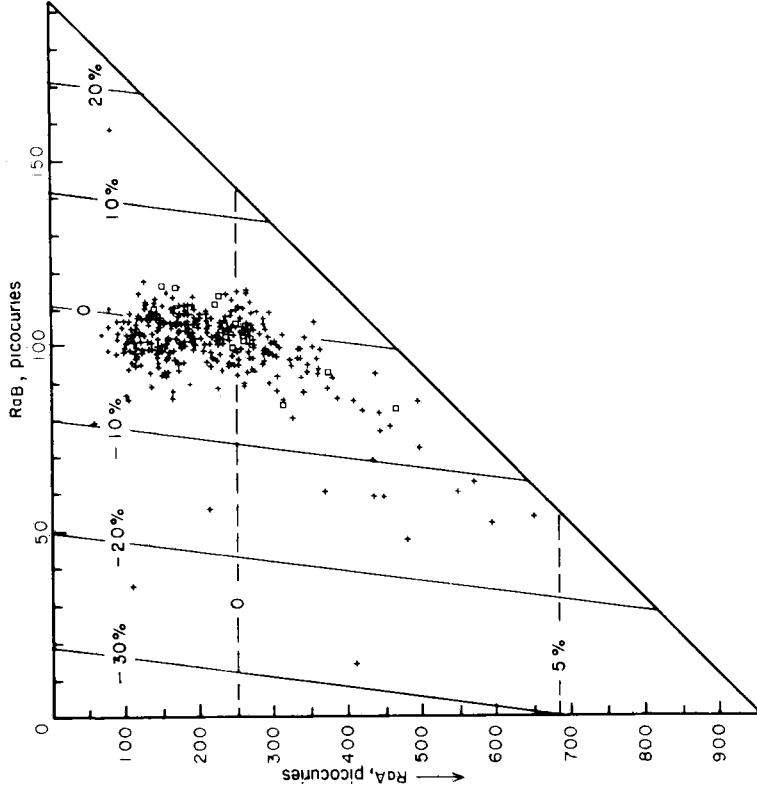


FIGURE 5. - Distribution of RaA, RaB, and RaC in mine atmospheres and the inherent errors for gross alpha and gross beta measurements.

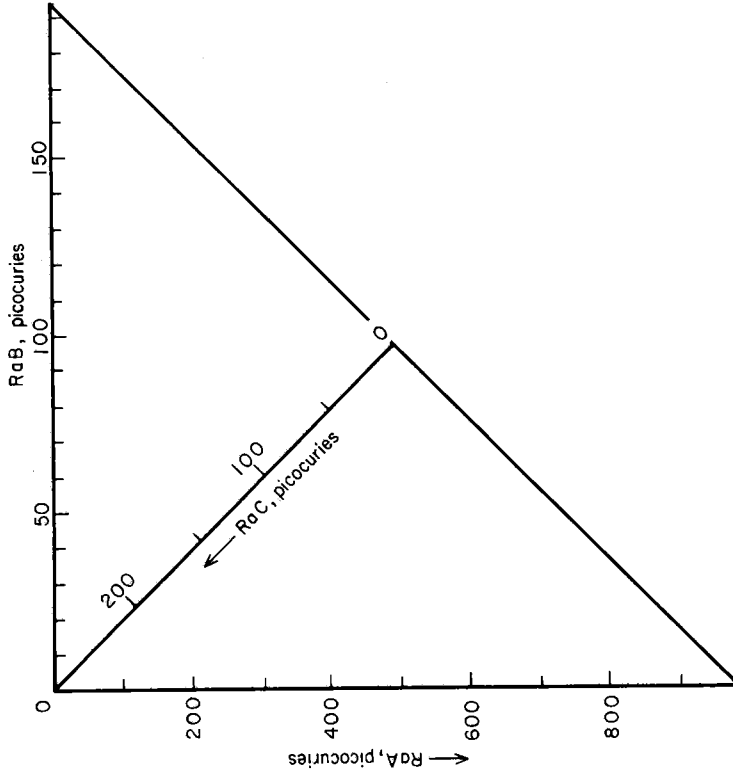


FIGURE 4. - Triangular graph for displaying distributions of airborne mixtures of RaA, RaB, and RaC.

One of the simplest methods of calculating inherent error is based upon the equilibrium filter paper activities determined by equations described by Evans (1). A constant supply of daughter products having an equivalency of one working level is assumed. Table 1 shows the calculated errors for pure RaA, RaB, and RaC based upon a zero inherent error for the test chamber mixture. Using this information, the isoerror lines shown in figure 5 as solid lines can be determined.

TABLE 1. - Filter equilibrium beta activity for pure RaA, RaB, and RaC and for test chamber mixture at 1.0 liter per minute and 1.0 working level

Daughter product	Picocuries	DPM ¹	Deviation from chamber mixture, percent
RaA.....	8,416	18,684	-28.76
RaB.....	14,991	33,281	+26.95
RaC.....	7,496	16,640	-36.52
Test chamber.....	11,809	26,216	0

¹Disintegrations per minute.

A good estimate of the probable inherent error limits can be made for the continuous beta method by selecting isoerror lines that include most of the data points shown in figure 5. A value of ± 8 percent includes most of these points.

The inherent error for the gross alpha method of continuous working-level measurements is calculated in the same manner as for the beta method. Table 2 shows the calculated errors for pure RaA, RaB, and RaC based upon a zero inherent error for the test chamber mixture that passes through the central portion of the field data.

TABLE 2. - Filter equilibrium gross alpha activity for pure RaA, RaB, and RaC and for test chamber mixture at 1.0 liter per minute and 1.0 working level

Daughter product	Picocuries	DPM ¹	Deviation from chamber mixture, percent
RaA.....	8,416	18,684	+8.84
RaB.....	7,496	16,641	-3.07
RaC.....	7,496	16,641	-3.07
Test chamber.....	7,733	17,167	0

¹Disintegrations per minute.

CALIBRATION OF DETECTORS

Both the alpha and the beta continuous working-level monitors are calibrated experimentally using the modified Tsivoglou method (10) for daughter concentration and working-level determinations. Calibrations using the radon test chamber are made during periods of daughter product stability with the

relative humidity in excess of 50 percent and the condensation nuclei in excess of 100,000 particles per cubic centimeter. A minimum of 3 hours' collection time elapses before the detector count rates are related to the working-level values, to allow daughter product equilibrium to be established on the filter paper. The beta detector requires a background measurement which is determined either at the end of the exposure and the decay time after removing the filter and shutting off the air supply or by use of a second detector with a known sensitivity relationship to the working-level detector. Operation of the detector at a fixed site minimizes problems with changing background values, and in most instances the background remains essentially constant throughout the measuring periods.

Airflow rates are carefully set at 1 liter per minute using a bubble meter to calibrate a flowmeter. The 1-liter-per-minute flow rate has been selected to maximize the filter life in a mining environment where the atmosphere has a high concentration of diesel combustion products and high humidity. Under these conditions, the detectors have been used continuously for 2-week periods without a significant increase in the filter resistance when fiber-glass filters are used.

To determine the calibration factor for a given detector unit, the average counts per minute (CPM) determined from the foregoing exposure techniques are corrected for dead time and background and then simply related to the average working level measured during the stable exposure period. A typical count rate for currently used beta detectors is 1,600 CPM per working level at an airflow rate of 1 liter per minute.

Direct count rate conversion to working levels assumes a steady state for the activity of RaA, RaB, and RaC being sampled. Such a condition can be achieved during calibration in a controlled test chamber. In a typical application, however, activity varies with time, and a direct conversion may be lower, higher, or equal to an "instantaneous" value for a given point in time. By integrating the counts over a time interval sufficiently long to account for the decay of all filter activity, through RaC, it is possible to arrive at an average working-level value for the time interval represented.

The total error of these methods of continuous working-level measurements would include (1) the inherent error, (2) the statistical error of the filter paper counting, (3) the statistical error of the background counting, (4) the error caused by plateout, (5) the error in the airflow rate, (6) errors caused by changes in self-absorption of the filter paper, and (7) errors of the working-level measurement made during calibration.

RESULTS AND DISCUSSION

Initial test work on both of the continuous working-level measuring methods was carried out in the Bureau of Mines radon test chamber at the Denver Mining Research Center. Not all of the typical mine atmosphere conditions can be simulated in this chamber, and additional testing has been conducted in the Bureau's experimental uranium mine near Uravan, Colo.

Figure 6 shows a plot of working levels and radon-222 measured over a 72-hour period. The working levels were measured with the beta detector unit,

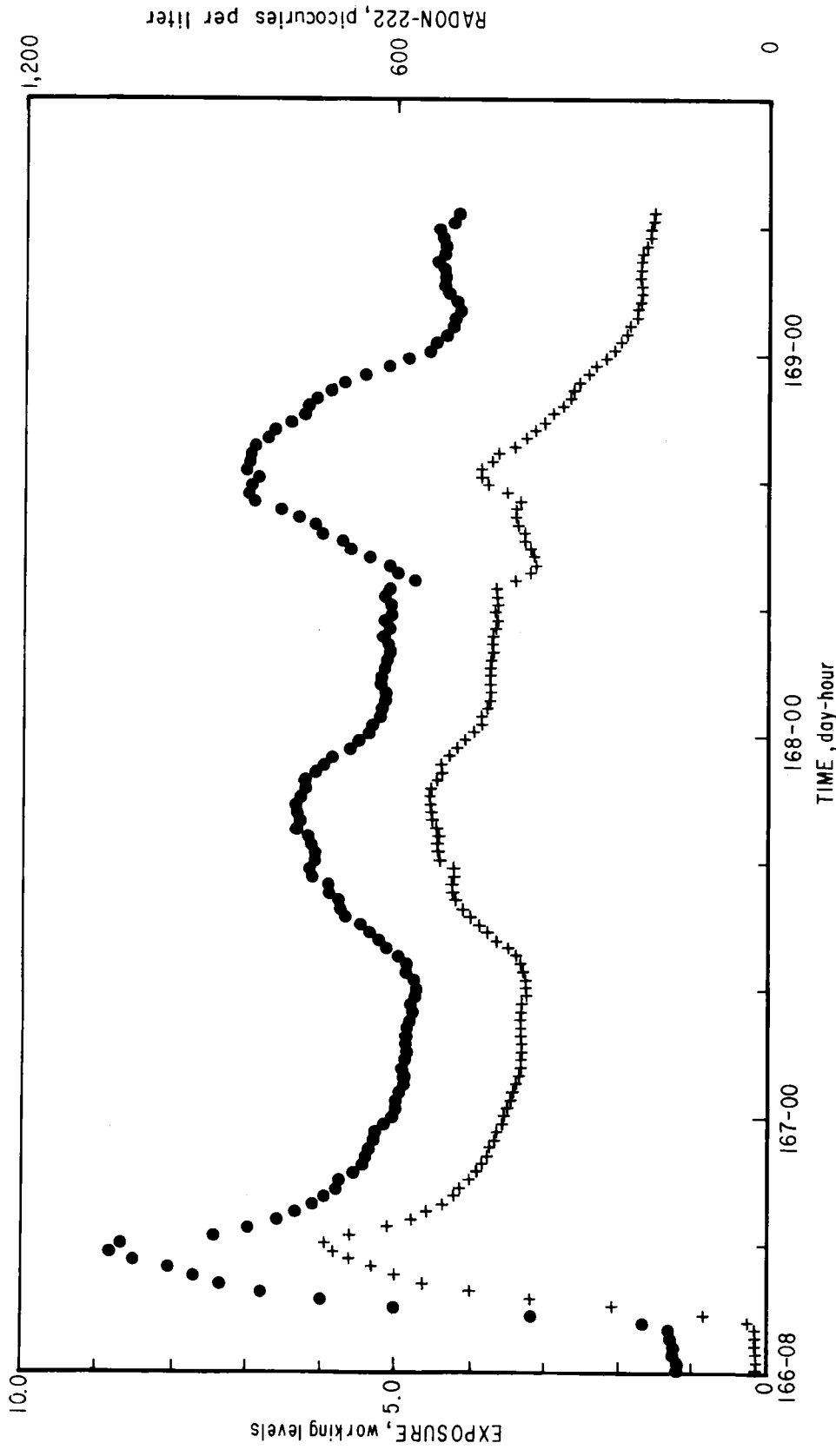


FIGURE 6. - Working-level exposures (plus marks) measured with beta detector and radon levels (solid circles) over a 72-hour period in Twilight mine.

and the radon was measured with a scintillation-type detector having a 500-cubic-centimeter volume. Both detectors were operated from the same controlled airflow system at 1.0 liter per minute.

At day-hour 166-09 both the radon-222 and the working level values were relatively low until a control bulkhead was closed and a reverse air-circulation fan was started at day-hour 166-11, resulting in a rapid buildup of atmospheric radionuclides at the air-sampling site. At the same time the fan was started, a diesel engine was placed in operation to simulate air conditions found in many active uranium mines. This raised the condensation nuclei level from less than 1,000 to over 200,000 particles per cubic centimeter. The reverse air circulation and condensation nuclei levels were then maintained until day-hour 168-09 while thermoluminescent (TL) working-level-hour-type dosimeters were being exposed. During this period the radon and working level values changed, primarily as a result of barometric pressure changes, as indicated by the rise in activity at day-hour 167-10 which followed a falling barometer.

During this period the TL dosimeters were exposed for 24 hours. Figure 7 shows a portion of this 24-hour exposure period with a plot of the

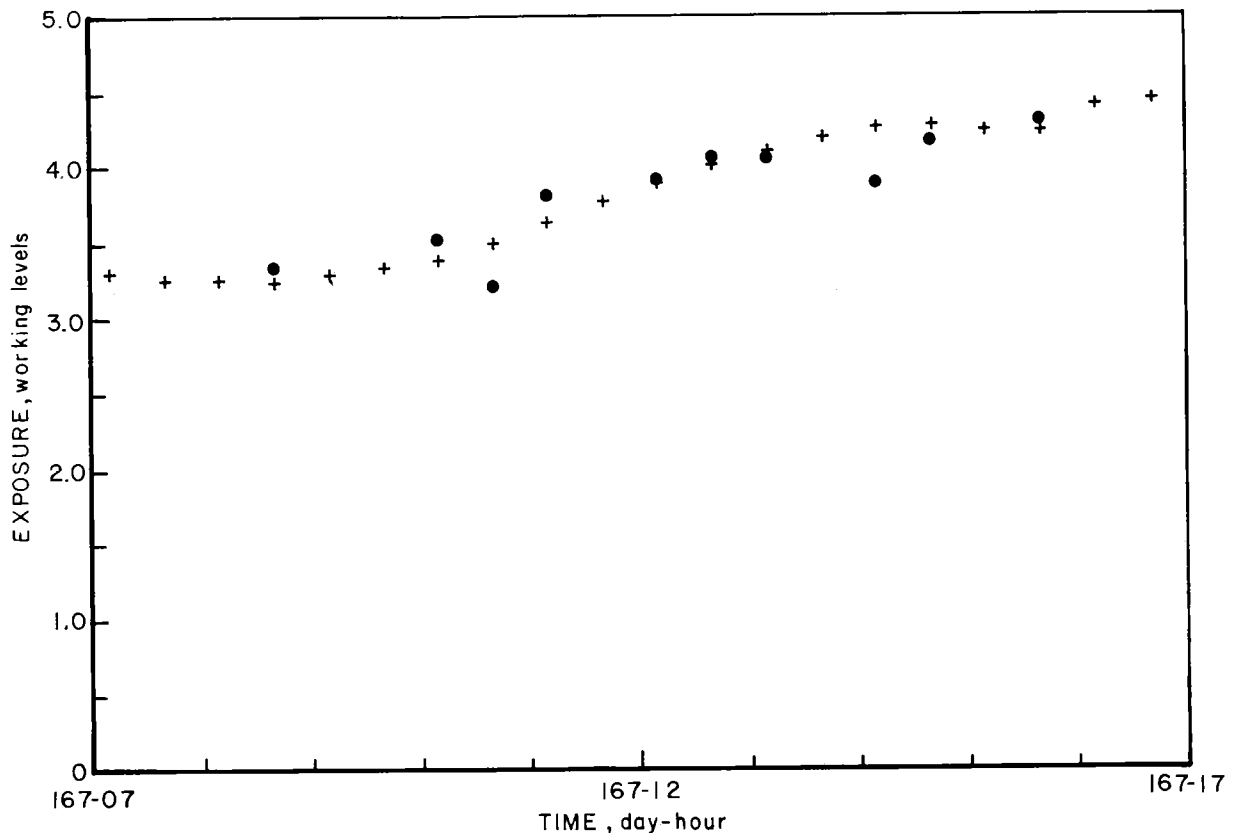


FIGURE 7. - Working-level exposures measured with the beta detector (plus marks) and by the modified Tsivoglou method (solid circles).

working-level values based upon the beta detector and a plot of working-level values measured by the modified Tsivoglou method (10). The scatter shown by the latter method is largely attributable to the usual variations in the working level that are integrated smoothly by the continuous methods of air sampling.

At day-hour 168-09 the diesel engine was stopped and the second stage of the recirculation fan was started. This resulted in a drop in condensation nuclei and a change in atmospheric radioactivity at the sampling site. Figure 6 clearly demonstrates that the working level and the radon level are not always directly related.

Also evident is a change in the working-level ratio⁶ after the diesel engine was stopped at day-hour 168-09. The most likely cause for this change is that the reduction in condensation nuclei in the mine atmosphere resulted in an increase in plateout of RaA (11).

Figure 8 shows the working-level exposure value measured with a continuous alpha detector and the radon-222 level for about a 25-hour period in the

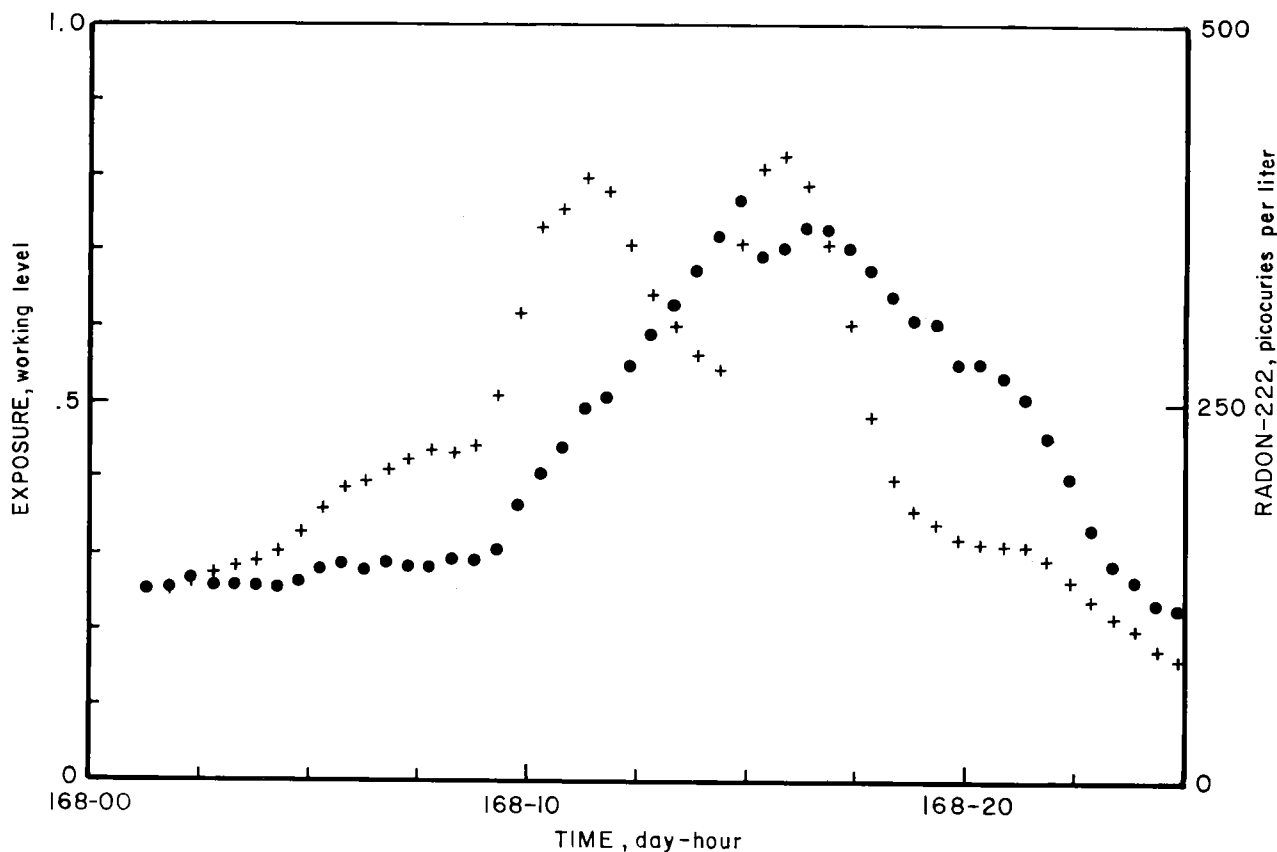


FIGURE 8. - Working-level exposures measured with an alpha detector (plus marks) and radon levels measured with a scintillation detector (solid circles).

⁶A working-level ratio is 100 times the working level divided by the radon concentration in picocuries per liter. It expresses the degree of equilibrium between radon and its daughters.

Twilight mine instrument room. The room is located underground near an air control bulkhead which has a relatively high level of airborne radionuclides on the side away from the room. Some of this radioactivity was leaking around the bulkhead and subsequently contaminated the fresh air supplied to the room.

The laboratory and field tests made in this investigation show that either alpha or beta detectors can be used in continuous working-level monitoring systems. However, under conditions where the condensation nuclei concentrations are less than about 40,000 particles per cubic centimeter, daughter product plateout becomes a problem with the alpha detector holder. Relatively low concentrations of condensation nuclei result in increased amounts of unattached daughter products (5, 7). These unattached radionuclides are collected on the surfaces of the air openings leading to the filter rather than on the filter surface itself, causing significant errors in the measurement.

The need to keep the distance between the detector and the filter paper as close as practical, when using an alpha detector, makes it difficult to design a holder that overcomes the plateout problem. The beta detector assembly described in this report avoids the problem by using an open-face filter holder with the detection of beta particles made from the backside of the collection filter.

In addition to the problem with plateout experienced with the alpha detector holder, there is a problem with filter papers. A membrane-type filter provides the least amount of alpha energy degradation, but it becomes clogged in a few hours at a flow rate of 1.0 liter per minute in a typical underground mine atmosphere. The fiberglass filter does not have this severe clogging problem, but it does have significant changes in its self-absorption characteristics for alpha particles. Test chamber studies show a 6-percent increase in self-absorption with changes in relative humidity from about 15 percent to about 70 percent.

SUMMARY AND CONCLUSIONS

Laboratory and fieldwork reported in this investigation show that continuous working-level measurements can be made using either alpha or beta detectors. The method requires a constant airflow rate and the collection of radon daughters on suitable filter papers. The inherent error analysis shows, by means of a graph, the probable inherent error for both alpha and beta measurements based upon typical underground mine atmosphere conditions.

Application of continuous working-level monitors in the Bureau's experimental mine shows the relationship between radon and working levels with changes in the characteristics of the mine atmosphere.

The use of alpha detectors in continuous working-level monitors is limited by problems with plateout of daughter products and self-absorption in the collecting filter. Measuring beta particles overcomes these problems, making this approach the more useful.

Continuous working-level monitors can be applied to control of radiation exposure in underground openings such as mines and caves, as well as in uranium ore processing mills and at mill tailing sites. In addition, the method has proven to be useful in the calibration of personal dosimeters designed to measure working levels over extended periods. By taking the integral of the activity over long sample periods, this method accounts for short-term changes of radon daughter product concentrations, resulting in exposure measurements equal or superior to "grab sample" techniques in overall accuracy.

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